Mixed Crystals in Semiconductors*

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1 Introduction

From both fundamental or academic and industrial interest, solid state physics of a mixed crystal of semiconductor has extensively been invetigated, which is prepared by alloying or making solid solution of two constituent semiconductors. Mixed crystals are also called as, simply, alloy or solid solution. One of the motivations is that by alloying two semiconductors one can change several semiconductor parameters from those of original semiconductors, for example, lattice constant, band gap, effective mass, activation energy of donors or acceptors. By studying the variation of the energy gap and effective masses with composition one can often get information about the band structures of the two components of a mixed crystal, and in some cases also even unknown parameters of the constituent semiconductors.

For the electronic devices, the mixed crystals are useful, because the continuous variation of parameters makes possible the design of a material with any gap width within the range covered by the compounds. This designability is especially useful for optical devices, such as infrared sensors, injection lasers, and electrolumicescence.

Since the study of Ge–Si mixed crystals of IV–Group in the periodic table, a variety of compound semiconductors have been reported, such as III–V, II–VI, and IV–VI Groups and if possible, between the different groups. Generally, if two semiconductors have approximately similar band structure, and if the lattice constants do not differ appreciably, they may easily become solid solution, or alloy. In this case, the parameters of a mixed crystal change continuously between those of the constituent semiconductors. As will be menioned later, in some mixed system, they become smaller than that of the compositions. That is, the parameters show minimum. However, if two compositions have different band structure, *i. e.*, different position of the conduction band in the wave vector space, there appears a "knik" in the parameters at some mole fraction.

Further, in the study of the mixed semiconductors, emphasis has also been placed on the scattering mechanism of the carriers, in addition to the dependence of a number of parameters on the mole fraction. As is well known, the scattering mechanisms in semiconductor are the lattice vibration (deformation potential, optical, piezoelectric), ionized impurity, neutral impurity, and dipole scattering. In the mixed crystal, however,

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it is one problem whether there is an additional scattering of the carriers caused by the distortion of the periodic lattice potential, which is known as alloy scattering.

We would like to review what kinds of mixed system have been studied so far, and to survey some of the interesting results. There are a variety of experimental methods; optical technique such as absorption, reflection, and emission spectrum, electrical one such as thermal and electrical conductivity, Hall effect, magnetoresistance and analytical one such as X-ray diffraction and electron paramagnetic resonance. The states of a sample are single crystal, powder, sintered specimen, and evaporated thin film. Preparation of a mixed crystal requires a great deal of skill. The imperfections may easily be introduced into the crystal lattice, and they may become scattering center of the carriers, trap or recombinnation center, which in turn give rise to a cause of degradation of electrical and optical properties. Although the crystal growth techniques are important, here we do not take up them. There are many review papers for the band structare of semiconductors[1], III-V groups[2,3], II-VI group[4], and general.[5,6]

2 Mixed Crystal System

Table I shows systems of the typical mixed crystals in semiconductor including in part semimetals. Here $A_{1-x}B_xC$ is expressed by (A,B) C and $AB_{1-x}C_x$ by A(B,C), where x is a mole fraction or atomic percent, and otherwise by AB-CD. As indicated later, the system having alloy scattering and "one-mode" or "two-mode"-type of the optical phonon mode are also indicated. The detailed discussions will be omitted here, but we shall be mainly concerned with the variation of the band gap in several groups as follows.

A) IV Group. As is well known, minima of the conduction band of Ge and Si are in the direction <111> and <100> in the wave vector space, respectively. We first look upon one typical example which shows a "knik" in the parameter upon alloying when the two components have different band structures. Figures 1-1 and 1-2 show the dependence of the energy gap E_g and electron mobility μ on the mole fraction. Experiment shows transition from a behavior of Ge-like <111> structure to that of Si-like structure. In this figure, the mobility μ is obtained by subtracting the lattice scattering and impurity scattering from the observed mobility. In this system, specific heat,[4-3] thermoelectric power,[4-4] ultrasonic attenuation[4-5] and phonon spectrum[4-6] have been studied.

B) III-V Group. This system is by far the most extensively investigated so far, from academic and applicational aspects. In Table II, a few parameters for this group are compiled; E_g,[5] electron effective mass,[5] and lattice constants.[3]

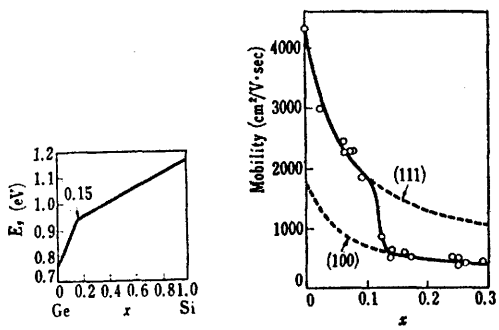

Fig. 1 The band gap E_g[4-1] and the electron mobility μ[4-2] against x for $Ge_{1-x}Si_x$.

Table I. Mixed Crystals in Semiconductors.

Ⅳ : Ge-Si[A-1)-5)a,c]	
Ⅲ-Ⅴ : In(As,P)[B-1)], In(As,Sb)[B-2)] (In,Ga)As[B-3)a], (Al,Ga)As[B-4)], (Al,In)As* Ga(As,P)[B-5)c], Ga(Sb,As)[B-6)b] (Al,Ga)Sb[B-7)], (Al,In)Sb[B-8)], (Ga,In)Sb[B-9)a] (Al,Ga)P[B-10)], (Ga,In)P[B-8)] InP-GaSb[B-8)], InP-GaAs[B-8)]	Ⅱ-Ⅵ : (Cd,Zn)S[C-1)b], (Cd,Mn)S[C-2)a] (Cd,Zn)Se* (Cd,Zn)Te[C-3)], (Cd,Mg)Te[C-4)] Cd(S,Se)[C-5)c], Cd(Se,Te)[C-6)], Cd(S,Te)* Zn(S,Se)[C-7)c], Zn(Se,Te)[C-8)b], Zn(S,Te)*
Ⅱ-Ⅵ : (Mercury Chalcogenide) (Hg,Cd)Te[D-1)], (Hg,Zn)Te[D-2)], (Hg,Mn)Te[D-3)] Hg(Se,Te)[D-4)] Hg(S,Se)[D-4,5)] Hg(S,Te)[D-4)]	Ⅳ-Ⅵ : (Pb,Sn)Te[E-1),4)], (Pb,Sn)Se[E-2),4)] Pb(Se,Te)[E-3)], Ge(Se,Te)[E-5)] PbSe-SnTe[E-4)], PbTe-SnSe[E-4)] PbTe-GeSe[E-6)], SnTe-GeSe[E-6)]
Ⅲ•Ⅴ-Ⅱ•Ⅵ : [F-1)] AlP-ZnS, AlSb-CdTe, GaP-ZnSe, GaAs-ZnSe, GaSb-ZnSe, GaSb-CdTe, InP-CdS, InP-CdSe, InAs-CdTe, InAs-ZnTe, InAs-HgTe, InSb-CdTe	
Ⅱ•Ⅵ-Ⅳ•Ⅵ : CdTe-PbTe[G-1,2,3)], CdSe-PbTe[G-1)]	
Ⅱ•Ⅵ-Ⅲ•Ⅵ : HgTe-In_2Te_3[H-1)]	Ⅱ-Ⅴ : Cd_3As_2-Zn_3As_2[I-1)]
Ⅱ-Ⅵ : Mg_2X, X=Ge, Si[K-1)], Sn[K-2)]	others: Bi_2Te_3-Tl_2Te_3[J-1)]

* : No data available to our knowledge.

a : Alloy scattering, *b* : "one-mode" type, *c* : "two-mode" type.

Al compounds are not so well studied because of difficulty of crystal preparation. III-V group semiconductors have a *zincblende cubic structure* and they may be well alloyed to each other. AlSb, GaP, and AlP have a band structure of <100> minima, while others have <000> minima. For the mixed system with different band structure, like Ge-Si, the "knik" in E_g is found, *i. e.*, for $GaAs_{1-x}P_x$ it occurs at x=0.44, and for Ga_{1-x} Al_xSb at x=0.7.

While for a system with the similar band structure, E_g varies monotonically with x; *e. g.*, for $InAs_{1-x}P_x$, E_g varies almost linearly with x. However, there are exceptions; *e. g.*, for $InAs_xSb_{1-x}$, E_g as well as m^* has a minimum at about x= 0.4-0.5, according to Coderre and Woolley, *et al*,[B-2)] although electron mobility varies linearly, as shown in Fig. 2. These systems are interesting in this sense. Many scientists pay attention to III-V group, because they are applicable for a semiconductor laser, Gunn effect, EL, and infrared detectors. For them all we are mainly concerned with a variation of parameters with the mole fraction including emission efficiency.

C) II–VI Group. For application to the optical devices, a great deal of research have also been made on this group. CdTe, ZnTe, and ZnSe have a *zincblende structure*, while CdS, CdSe, and ZnS a *wurtzite structure*. Apart from the detailed discussions, this group has nearly the same band structure of <000> minima. HgTe is also II–VI compound, but it is rather a semimetal. Later we will see Hg-chalcogenides.

Figure 3 shows the relation between the effective mass and band gap, which is compiled from the data obtained so far.[4] We can estimate m^* from this figure, if E_g is known. In fact, however, one has to take into consideration of the anisotropy of m^* and spin-orbit interaction for this relation. For simplicity, we have neglected them. Since the band structures of this group are similar to each other, there is no "knik" in E_g-x curve ; *e. g.*, for (Cd, Zn)S and Cd(S,Se) systems, E_g changes linearly with x. But for some examples such as $ZnSe_xTe_{1-x}$ and $Cd_{1-x}Mn_xS$, E_g

Table II. The band gap E_g,[5] electron effective mass m^*/m_0,[5] and lattice constant a_0[8] for III-V semiconductors.

	E_g(eV) 0 K	300K	m^*/m_0	a_0
InSb	0.235	0.18	0.0155	6.47
InAs	0.42	0.35	0.024	6.06
GaSb	0.81	0.7	0.042	6.09
InP	1.42	1.35	0.073	5.87
GaAS	1.52	1.4	0.08	5.65
AlSb*	1.60	1.52	0.39[+)]	6.13
AlAs	2.1-2.3		0.5-0.8[+++)]	5.66
GaP*	2.34	2.26	1.37[++)]	5.45
AlP*	2.6-2.7[B-10)]			5.46

*) Conduction band minima <100>, others <000>.

+) T. S. Moss and B. Ellis: Proc. Phys. Soc. (London) **83** (1964) 217.

++) H. C. Montgomery : J. appl. Phys. **39** (1968) 2002.

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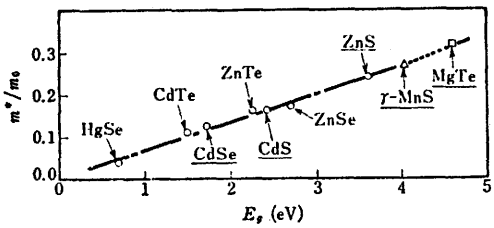

Fig. 3 Relation between the band gap and effective mass. "——" indicates the wurtzite structure, the others the zincblende structure. Only the band gap is known for MgTe (R.H. Bube: *Photoconductivity of Solids* (John Wiley and Sons, New York, 1960)).

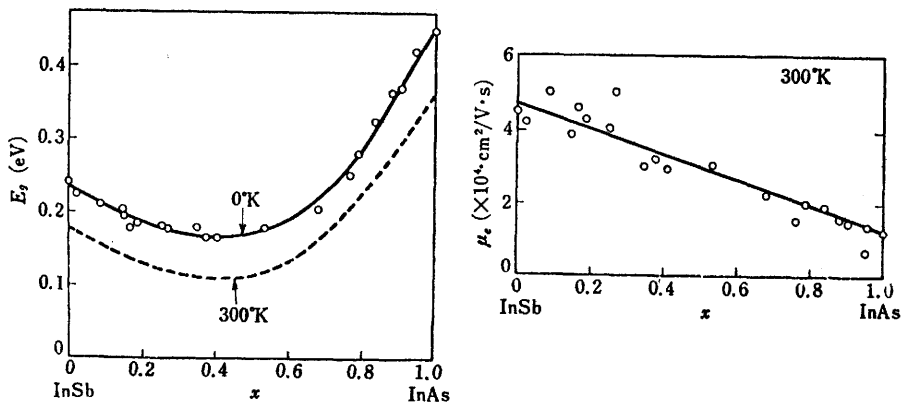

Fig. 2 The band gap E_g and the electron mobility μ_e against x for $InAs_xSb_{1-x}$, after Coderre, *et al.*[B-2)]

minima have been reported at x=0.64 (Larach, *et al*),[C-8] and 0.03 (Ikeda),[C-2] respectively. These minimum phonomena are not well understood.

One of the characteristic features of this group is so-called "self-compensation" of doping in crystals, and thus it is difficult to make a crystal with desired conductivity type, n- or p-type. At the same time, analysis of the measured conductivity or Hall effect is not so simple because of the presence of the lattice vacancies. That is why the data of transport phenomena are rather fewer than those of optical experiments. II-VI group, however, has a wide band gap, which benefits the electronic devices aimed for visible EL and laser, etc. The changes such as in the emission peak and emissivity have been studied.

D) II-VI Group; Mercury compounds. This group is interesting for the application to the infrared detectors and for an inversion of the band structure. Especially $Hg_{1-x}Cd_xTe$ has been extensively studied, which was taken up at the International Conference on Semiconductor at Kyoto, 1966.[D-1]

HgTe and HgSe, semimetal, have a crystal structure of zincblende type, just as CdTe and ZnTe. The conduction band minimum is of k= <000> type. Figure 4 indicates the band structure of HgTe, a mixed crystal with CdTe in the neighborhood of k=0, the change in the band gap $E_g(=E_{\Gamma_6}-E_{\Gamma_8})$, and m^*/m_0. At k=0 in the wave vector space, the light electron band Γ_8 and the heavy hole band Γ_8 overlap each other, and below them comes the light hole band Γ_6. The amount of the overlapping between electron and hole bands E_t is called as a thermal energy gap; at 4.2 K, E_t= 0.003−0.02 eV for HgTe and 0.07 eV for HgSe. Both E_g and E_t depend on pressure and temperature.[D-1]

Now, by alloying HgTe with CdTe, E_g reduces to zero at x=0.15, showing a linear variation, while m^* (**k-p** perturbation method by Cardona) becomes zero at this mole fraction. Temperature dependence dE_g/dT and pressure dependence dE_g/dP for HgTe are reported to be negative, but by alloying with CdTe the two coefficients become positive. This is considered due to the difference of the band structure. The similar inversion

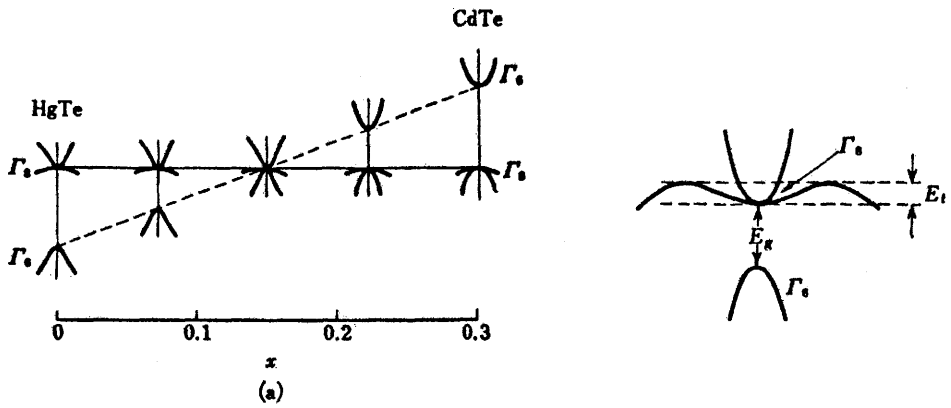

Fig. 4 (a) The variation of the band structure for $Hg_{1-x}Cd_xTe$(Wiley and Dexter[D-1]), and the band structure of HgTe (Harman[D-1]).

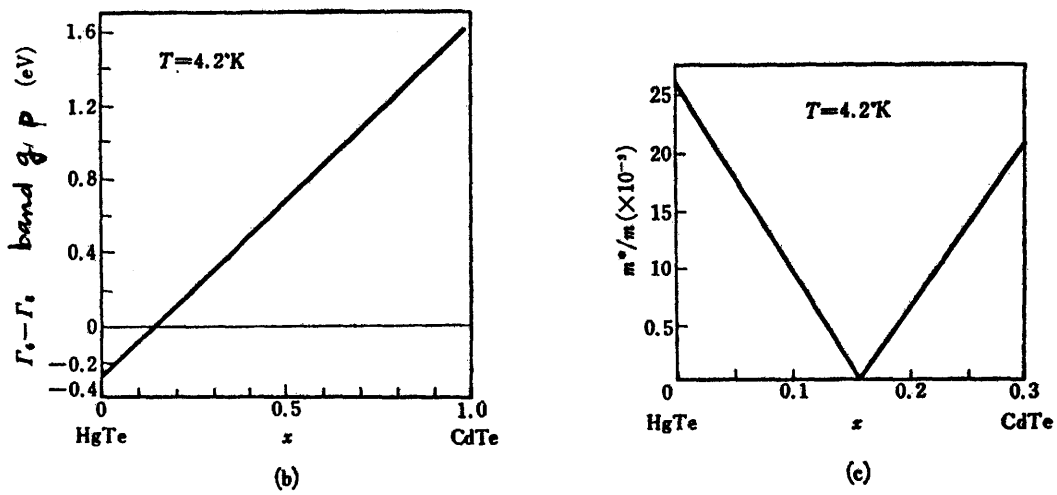

Fig. 4 (b) The band gap Γ_6-Γ_8 against x.
(c) The effective mass against x (M. Cardona : J. Phys. Chem. Solids **24** (1963) 1543.).

of the band structure is also found for (Hg,Zn) Te mixed crystal.[D-2] 'Mixed crystals among the mercury compunds Hg (S, Se), Hg (S, Te), and Hg (Se, Te), are observed to change in the thermal energy E_t; for instance, E_t vanishes to zero at x=0.18 for HgS_xSe_{1-x}.[D-5]

E) IV–IV Group. This group crystallizes in a NaCl type structure. In Table III, a direct band gap E_g and effective mass of both types of carriers are shown. The conduction band minimum lies at k=0, while the valence band maximum is along <111> direction, in addition to k=0, which form a many-valley structure.

Figure 5-1 shows a typical band structure for PbTe and SnTe,[5] and Fig. 5-2 gives the variation of $E_g(=E_{L_6^-}-E_{L_6^+})$ with x, obtained by Dimmock, *et al.* from the laser emission spectrum.[E-1] In this case, the two bands of L^+_6 and L^-_6 are inverted by alloying. It should be noted that $d|E_g|/dT$ for the PbTe-like band structure is negative and for the SnTe-like one positive. The similar band inversion and temperature dependence are found for $Pb_{1-x}Sn_xSe$ at x=0.15 (12K).[E-2] While for $PbTe_{1-x}Se_x$ no inversion is observed, E_g decreases monotonically with x.[E-3] As for the infrared laser

Table III. The band gap E_g, electron effective mass, and hole effective mass.[5]

	E_g(eV)		electron		hole	
	0 K	300K	$m_\parallel^*/m_0$	$m_\perp^*/m_0$	$m_\parallel^*/m_0$	$m_\perp^*/m_0$
PbS	0.29	0.42	0.105	0.008	0.105	0.075
PbSe	0.15	0.29	0.07	0.04	0.068	0.034
PbTe	0.19	0.32	0.24	0.02	0.31	0.024
SnTe	0.3	0.18				
GeTe						

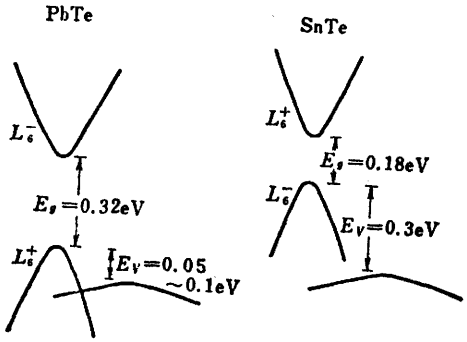

Fig. 5 - 1 The energy band (300K) for PbTe and SnTe (L. M. Rogers: Brit. J. appl. Phys. **1** (1968) 845.).

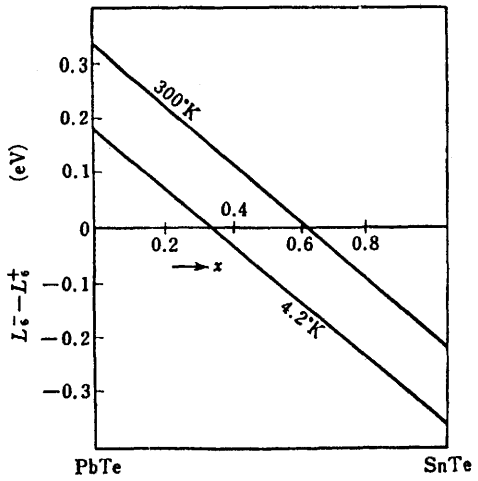

Fig. 5 - 2 The band gap agaist x for $Pb_{1-x}Sn_xTe$ (Dimmock, *et al.*[E-1]).

material, these materials are useful, and radiation wavelength of 15 μ and 31 μ are measured for (Pb, Se) Te,[E-1] and (Pb,Sn) Se, respectively.[E-2]

Recently GeTe has been paid attention as a degenerate "semiconducting superconductor," which has a many-valley band structure like SnTe.[6] The critical temperature T_c is dependent on the carrier (hole) concentration, and they become superconductive state at more than $p=8\times10^{20}$ cm^{-3}. T_c is a little higher for GeTe than for SnTe, but even in this case T_c is less than 0.43K. We believe that the study of superconducting properties for a mixed crystal of (Ge, Sn)Te may be interesting, though no experiment has been reported. At the same time, it may be interesting to study a new material such as SiS, SiTe, SiSe, particularly concerning with the band structure.

F) Others. We have talked here about a mixed crystal made of the same group. In Table I we have given some mixed systems made of the inter-groups. III-V-II-VI systems are investigated to a large extent by the Soviet scientists.[F-1] In general, for the II-VI-IV-VI and II-VI-III-VI systems, semiconductor parameters have the "knik" like Ge-Si system, because the crystal structure or band structure are different. There are some systems which show a change in the conductivity type; *e.g.*, in $Cd_{3-x}Zn_xAs_2$ a p-type conductivity changes to an n-type at a certain value of x.[I-1] It is likely that to grow a homogeneous crystal for these systems is far more difficult.

3 Mobility and Phonon Spectrum

In addition to a change in the band gap, as we have surveyed henceforth, some new phenomena are found solely for a mixed crystal, which are not observed in a constituent semiconductor.

Firstly, the carrier mobility is also determined by an alloy scattering as well as the

usual scattering mechanism, as[7]

$$\mu_A = \frac{(2\pi)^{1/2} e\hbar^4 N_a}{3(kT)^{1/2} m^{*5/2} (E_1 - E_2)^2 x(1-x)} ,$$

where $E_1 - E_2$ is the energy difference between the band gaps of the alloy components, N_a the density of lattice sites ($\sim 2\times 10^{22} cm^{-3}$), and x the mole fraction, respectively. Some mixed crystals are reported for the scattering to be effective; Ge-Si, (In,Ga)As, (Ga,In)Sb, and (Cd, Mn)S. In fact, it is difficult, however, to discriminate it from the observed mobility, since the crystal growth without lattice vacancies or inhomogeneities is considerably difficult.[C-4] And the $T^{-1/2}$ dependence of the mobility which is derived by subtraction of the lattice and impurity scattering mobilities from the measured ones, is not only determined by the alloy scattering, but also by such as disorder or imperfections involved in the crystal.[7]

Secondly, another problem to be noted in the physics of the mixed crystal is the optical phonon spectra in the long wavelength limit. Recently Chang and Mitra have compiled the experimental data to date,[8] and noticed that so far as the behavior of the long wavelength optical phonons is concerned, there exist two types of mixed crystals; "one-mode" and "two-mode" type. In the one-mode type, optical mode frequencies (infrared or Raman-active) vary approximately linearly and the strength of the mode remains nearly constant. Examples of this behavior are $Na_xK_{1-x}Cl$, $Ni_xCo_{1-x}O$, KCl_xBr_{1-x}, $K_xRb_{1-x}Cl$, $(Ca, Ba)_xSr_{1-x}F_2$, $GaAs_{1-x}Sb_x$, $Zn_xCd_{1-x}S$, and $ZnSe_xTe_{1-x}$. In the two-mode type, there appear two modes in the neighborhood of those of the end members. In addition, the strength of each phonon mode of the mixed crystal is approximately proportional to the mole fraction of the component it represents. Examples of the two-mode type of behavior are $InAs_xP_{1-x}$, Ge_xSi_{1-x}, $GaAs_xP_{1-x}$, CdS_xSe_{1-x}, and ZnS_xSe_{1-x}.

In Fig. 6 is shown the behavior of long-wavelegth optical phonons as functions of

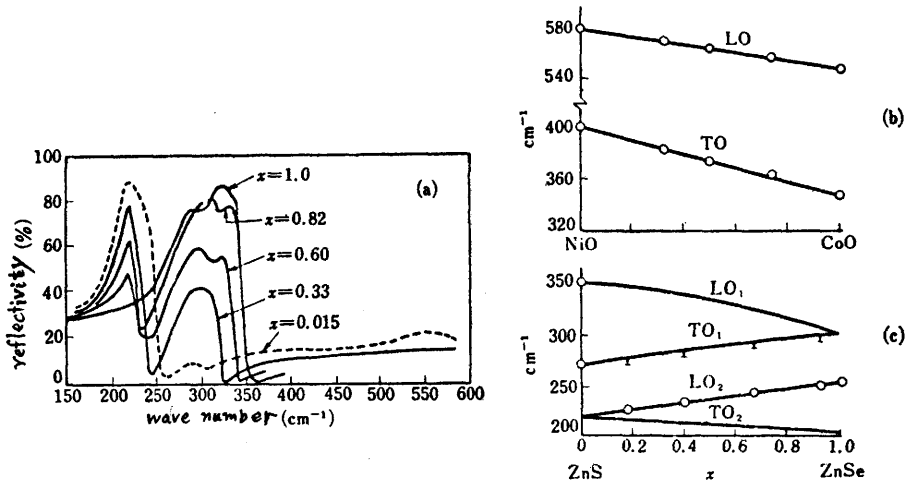

Fig. 6 (a)The optical reflectivity against the wave number for $ZnS_{1-x}Se_x$. (b) The "one-mode" type of $Ni_{1-x}CoxO$, and (c) the "two-mode" type behavior of $ZnS_{1-x}Se_x$.

mole fraction. The top part of the figure illustrates the behavior of $ZnS_{1-x}Se_x$, which belongs to the two-mode class, while Fig. 6 (b) illustrates the behavior of $Ni_{1-x}Co_xO$, which belongs to the one-mode, and Fig. 6 (c) does the concentration dependence of optical modes for the former.

According to a simple model of the lattice vibration for a diatomic crystal, AB, a transverse optical phonon frequency ω_T in the long wevelength limit is given by the reduced mass μ_{AB},

$$\omega^2_T=2C\left[\frac{1}{M_A}+\frac{1}{M_B}\right]=\frac{2C}{\mu_{AB}} ,$$

where M_A and M_B are the masses of the individual atom, and the constant C the force constant between them. The longitudinal optical frequency ω_L, however, is not written simply like this. But there is well-known "Lyddane-Sachs-Teller" relation given by

$$\varepsilon(0)/\varepsilon(\infty)=\omega^2_L/\omega^2_T ,$$

where $\varepsilon(0)$ and $\varepsilon(\infty)$ are the static and optical dielectric constant, respectively. This relation is well established for the ionic crystals such as alkali halides and GaAs. Now if we assume a mixed crystal $AB_{1-x}C_x$ to be the one-mode type, then it can be treated by the virtual crystal model, where one replaces the atoms B and C with a virtual atom such that the reduced mass μ of the primitive unit cell is given by

$$\mu^{-1}=M_A^{-1}+(1-x)M_B^{-1}+xM_C^{-1},$$

or

$$\mu^{-1}=M_A^{-1}+\frac{1}{[(1-x)M_B+xM_C]} .$$

Thus ω_T depends on the mole fraction. Chang and Mitra have derived a simple criterion for the prediction of one- or two-mode type of behavior of a mixed crystal, taking into account of the change in dielectric constant, polarization effect, and randomness of atomic distribution. A two-mode (one-mode) type of mixed crystal of the type $AB_{1-x}C_x$ must (not) have one substituting element whose mass is smaller than the reduced mass of the compound formed by the other two elements. If

$$M_B<\mu_{AB}<M_A,\ M_C$$

then there occurs a local mode near $x=1$ and 0, which belongs to the two-mode type. For instance, for $GaP_{1-x}As_x$ of this type

$$M_P(=31)<\mu_{GaAs}(=36.4)<M_{As}(=74.9),\ M_{Ga}(=70).$$

On the other hand, for $GaAs_{1-x}Sb_x$ of the one-mode type

$$M_{Ga}(=70),\ M_{As}(=74.9)>\mu_{GaSb}(=44.3) ,$$

$$M_{Sb}(=121.8)>\mu_{GaAs}(=36.1).$$

We can apply this criterion to any mixed crystal in order to know whether it is of the one- or two-mode type.

4 Summary

We have surveyed the kinds of mixed crystal of semiconductor, which have been compiled from data reported to date, and we have reviewed some of the interesting

results. As far as the variation of the band gap is concerned, it is summarized that by alloying two semiconductors, there are four kinds of variation : (i) linear change, (ii) nonlinear, (iii) occurrence of a "knik" at some concentration, and (iv) appearance of minimum. Further, phenomena characteristic of mixed crystals are the alloy scattering and existence of two kinds of the long-wavelength optical modes. These facts may be considered to be reflected from the crystal structure and band structure.

The study of mixed crystals has not only been made on semiconductor, but also on ionic crystal, dielectrics, magnetic material, superconductors, and over a variety of the fields of solid state physics. For further understanding of physics of the mixed crystals, both theoretical and experimental studies are required.

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III-V Group

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